

# **Identification and Quantification of the Temporal and Spatial Scales of Variability in Particulate and Dissolved Material Associated with Specific Land-use Activities in the Penobscot River System**

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## **LONG-TERM GOALS**

The long-term goals of this project are to identify and quantify specific optical and chemical characteristics of the colored particulate and dissolved fractions of the Penobscot River water system that are associated with defined land use activities (land use proxies), and to determine the scales of variability over which these proxies can be detected both temporally (i.e. seasonal and episodic events) and spatially (from the source into coastal waters).

## **OBJECTIVES**

Our main objectives are to:

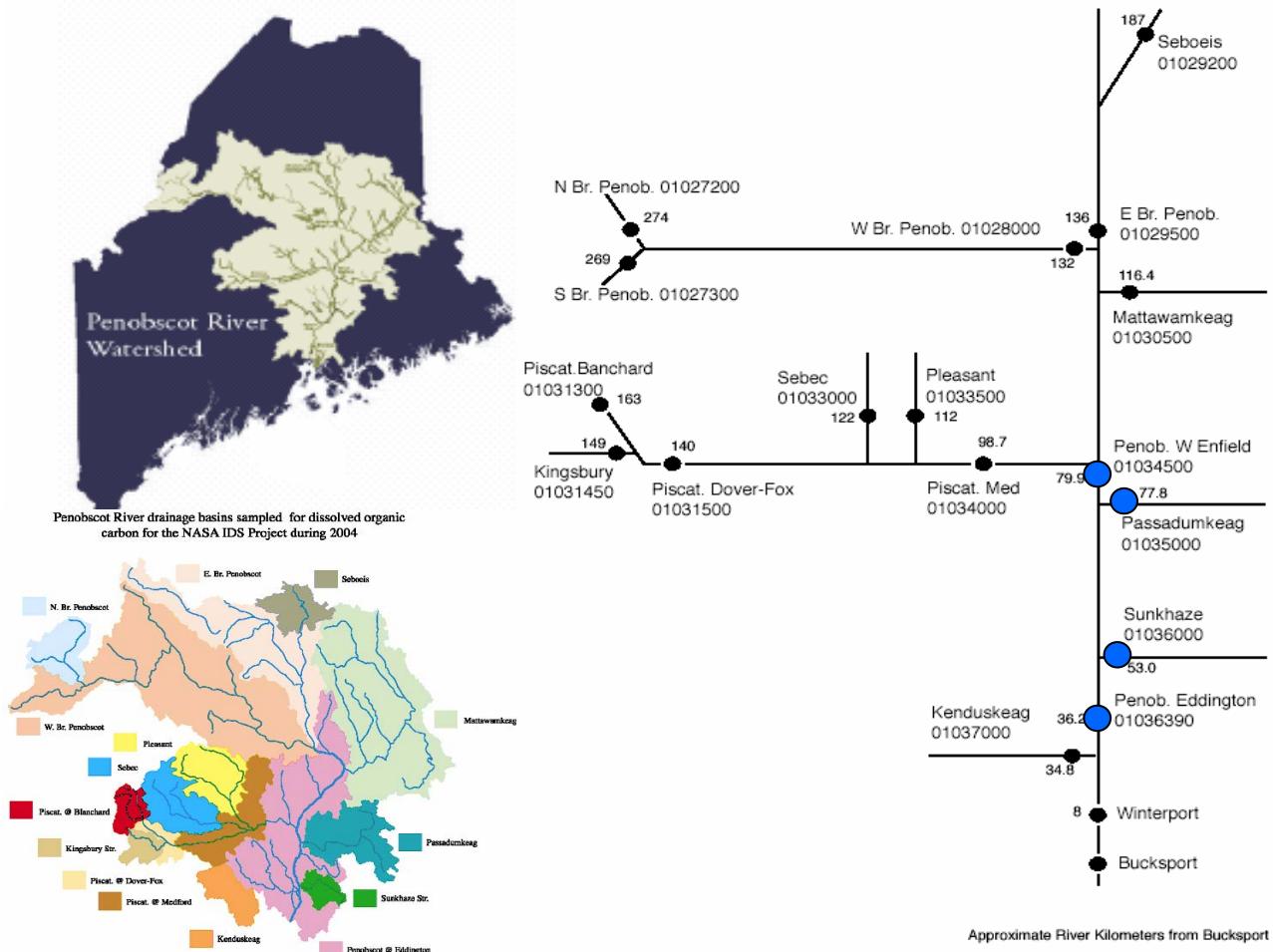
- develop optical proxies for biogeochemical properties (i.e. algal pigments, total suspended solids, particulate and dissolved organic carbon) that can be applied to in situ optical observations to obtain highly resolved temporal estimates of those biogeochemical properties
- identify optical proxies for land use activities (e.g. specific fluorophores linked to agricultural activities or wetlands)
- quantify the time and space scales for which these proxies behave as conservative tracers in the Penobscot River, Penobscot Bay and ultimately in the coastal waters of the Gulf of Maine.

## **APPROACH**

Penobscot River Watershed Sampling -We conduct approximately monthly sampling of nearly 20 stations within the Penobscot River Watershed (Figure 1). Water samples are collected and discrete optical observations are made with a WET Labs Triplet sensor (chlorophyll fluorescence, CDOM fluorescence and backscattering). Water samples are analyzed for chlorophylllic pigments (Yentsch and Menzel 1963), 3d excitation emission matrices (Coble 1996), particulate and dissolved absorption (with the particulate component separated into algal and non-algal components), particle size distributions (LISST; Agrawal and Pottsmith. 2000), particulate organic carbon and nitrogen and

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inorganic nutrients (CHN and  $\text{NO}_3$ ,  $\text{NO}_2$ ,  $\text{SiO}_2$ ,  $\text{PO}_4$ ; UC Santa Barbara routine chem lab), dissolved organic carbon and total dissolved nitrogen (DOC and TDN, Dr. George Aiken, USGS Boulder; Aiken 1992); total suspended solids and volatile solids (TSS and %VS; US EPA 1979), pH, conductivity. Optical proxy relationships are developed from the paired optical biogeochemical properties for application to moored optical observations (see below).



**Figure 1. Map of Maine with the Penobscot River Watershed highlighted, B. Major tributaries and drainage basins, each dominated by a specific coverage type (e.g. bog, agriculture, forest). C. Monthly sampling stations within the watershed (USGS number and km from Penobscot Bay indicated). Moored sensors deployed at West Enfield, Sunkhaze Stream Passadumkeag River and Eddington (blue symbols).**

Penobscot River In Situ Optical Observations - WET Labs Triplet sensors are deployed at four stations within the river (Figure 1C). These sensors are located at two locations within the Maine Branch at West Enfield and Eddington and at the mouth of two highly colored tributaries (Passadumkeag and Sunkhaze Stream) so as to investigate how well a highly colored and distinct optical input can be detected upon dilution into the main branch and also to investigate the conserved behavior of colored (and presumably carbon-rich) matter in the river. Optical proxy relationships are applied to these in situ optical observations to develop biogeochemical time series in the river.

Penobscot Bay Surveys -We have developed, constructed and deployed a ship-based observational system on a local research vessel (ARGO Maine) that routinely travel through the Penobscot River System from Bangor to the coastal waters. The operator turns on the flow-through system whenever the ship is in operation. This system yields GPS location, surface (~3m depth) temperature, salinity, single-wavelength beam attenuation, and chlorophyll and CDOM fluorescence with approximately 8 m resolution. Additionally, we conduct complete and focused surveys of Penobscot Bay twice per year with the goal to elucidate the fate and transport of particulate and dissolved matter from the River to the coastal waters. We employ a variety of optical platforms in addition to the flow through system (i.e. profilers, AUV, drifters) and collect discrete water samples for biogeochemical analysis. These data complement the optical proxy database.

Penobscot Bay and Gulf of Maine Moored Optical Observations - We have augmented the existing 3m optical packages on GoMOOS moorings E and I (downstream and upstream of Penobscot Bay in the Eastern Maine Coastal Current) with CDOM fluorometers to complement the multispectral IOP and AOP packages (ac-9, vsf3, chlorophyll fluorometers, 7-channel downwelling irradiance and upwelling radiance). We have also instrumented mooring F in the Penobscot Bay with a WET Labs FLNTU sensor to monitor chlorophyll and CDOM fluorescence and backscattering sensor as well as 7-channel incident irradiance and upwelling irradiance. The GoMOOS program provides real time hourly observations. Water samples are collected from these three moored sites during deployment/recovery cruises. They are analyzed as above. These data complement the optical proxy database.

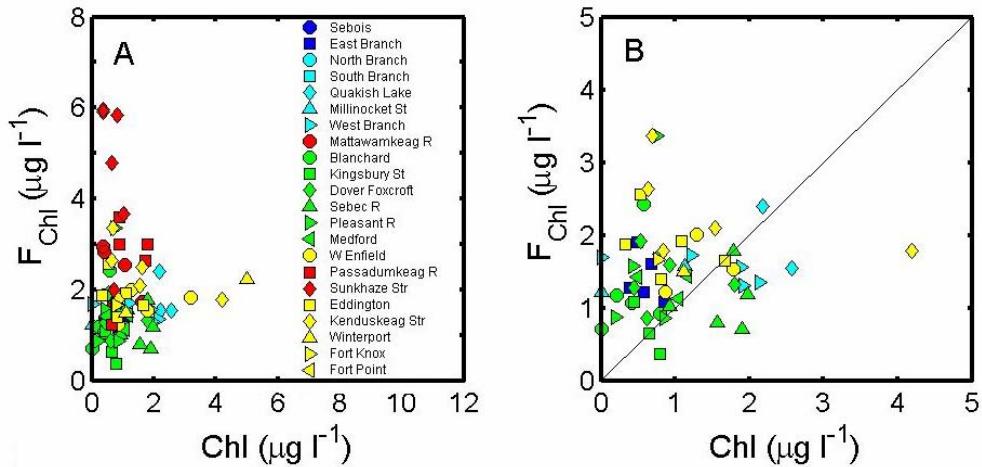
## WORK COMPLETED

We have conducted nearly monthly sampling of the watershed stations since May 2004. All optical data have been processed. All biogeochemical data have been processed or are currently being analyzed by external laboratories. Optical proxies have been generated from this data set and applied to the 2005 moored optical time series. We have conducted three surveys of Penobscot Bay (May 2005, October 2005, June 2006) and have one scheduled for October 2006. We have begun analyzing the GoMOOS time series observations to examine temporal coherence between river signatures and bay signatures. We have prepared a series of papers for the Ocean Optics Meeting in Montreal highlighting a range of aspects of this project.

## RESULTS

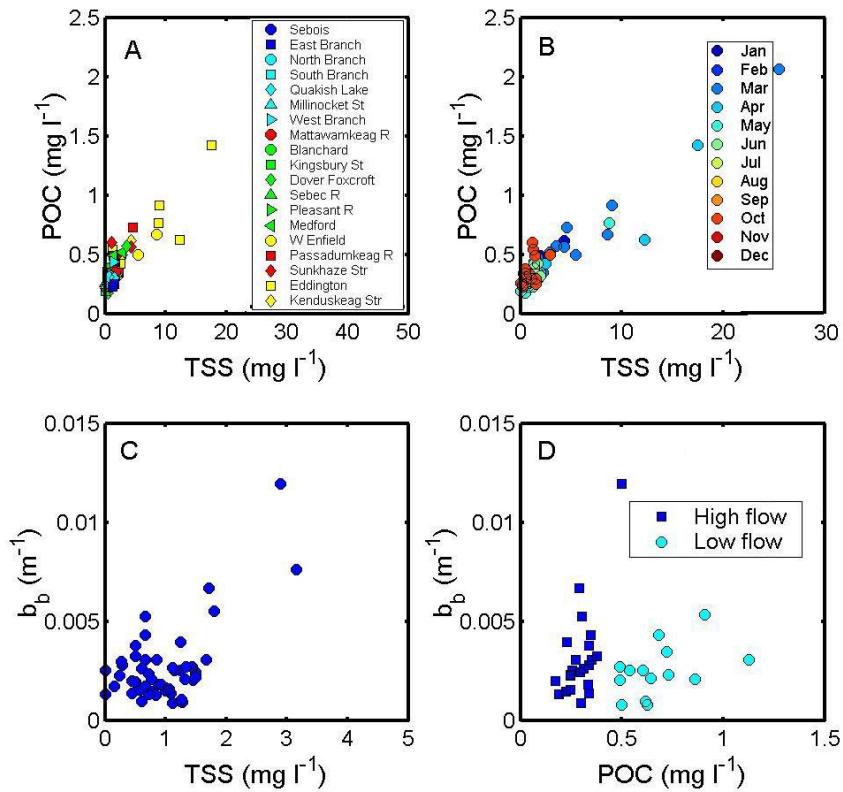
### Optical Proxies

Algal Concentration- Chlorophyll concentrations ranged from nearly zero to  $12 \mu\text{g l}^{-1}$ , with the lowest values occurring in the winter and in the agricultural subwatershed of the Piscataquis River tributaries. The highest values sampled were observed in 2004 in the area of the lumber mill in Millinocket along the West Branch of the Penobscot River, associated with an intense cyanobacterial bloom. These nearly annual blooms of cyanobacteria are thought to occur in response to release of phosphate by the mill. We observed a troublesome relationship between the calibrated in situ chlorophyll fluorometer observations and discrete chlorophyll concentration (Figure 2A). While most of the observations clustered in the 0 to  $2 \mu\text{g l}^{-1}$  range, one set of observations indicated high chlorophyll fluorescence in the absence of pigment, while the other indicated high pigment concentrations in the near absence of fluorescence. These are explained by considering the configuration of the chlorophyll fluorometer of



**Figure 2. Chlorophyll proxy derived from in situ chlorophyll fluorescence measured by the WET Labs triplet sensor vs. extracted chlorophyll concentration collected at discrete stations during monthly sampling program. A. All data, B. observations after high CDOM stations and cyanobacterial bloom stations were removed, line is one-to-one.**

the WETLabs triplet sensor. The chlorophyll fluorometer excitation is provided by a 470 nm LED; a long pass red filter on the detector provides detection centered at 695 nm. This excitation/emission pair is not tuned to detect chlorophyte algae or cyanobacteria and thus was insensitive to the large cyanobacterial bloom observed in the Western Branch of the Penobscot River in summer 2004. In addition this excitation/emission pair is sensitive to very high concentrations of fluorescing dissolved organic matter found in the tributaries that drain freshwater wetlands and bogs, which contaminates the signal. In the absence of those two scenarios, the relationship between calibrated in situ chlorophyll fluorescence and extracted chlorophyll concentration is accurate but very imprecise (Figure 2B), likely because of species- and physiologically-dependent variations (Marra and Langdon 1993). However, that said, the chlorophyll fluorometer does provide qualitative and comparative estimates of phytoplankton biomass with concentrations of the correct order.

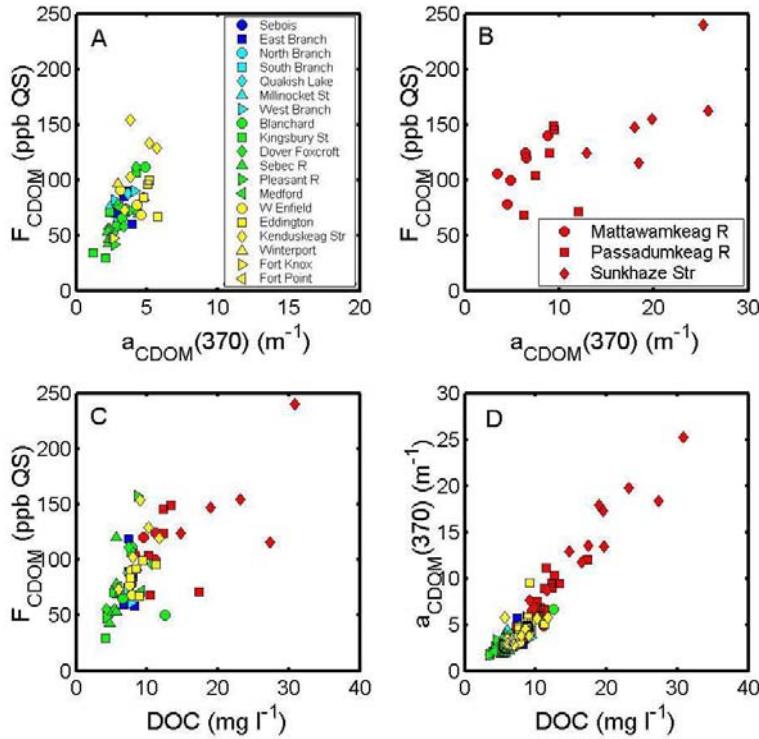


**Figure 3. Particulate organic carbon (POC) proxies. Relationship between POC and total suspended solids (TSS) by station (A) and month (B). Relationship between the in situ backscattering coefficient as measured with the WET Labs triplet and TSS (C) and POC (D) measured on discrete samples collected during the monthly sampling program. The backscattering POC proxy observations are distinguished by high and low flow regimes, see text for details.**

Particulate Organic Carbon - Particle concentrations, as quantified by both TSS and POC, ranged from nearly zero to approximately 25 (mg solids l<sup>-1</sup>) and 2.0 (mg C l<sup>-1</sup>), respectively (Figure 3A) and a strong correlation was observed between them for the freshwater stations. The highest values occurred during the high discharge period in the spring (Figure 3B). Unfortunately it was not possible to sample with the triplet sensor during these extremely high flow periods. However, linear relationships were observed between particle backscattering and both TSS and POC (Figure 3C and D), and thus the relationship observed at lower concentrations is extended to the higher concentration for two different discharge modes.

Dissolved Organic Carbon - The dissolved organic matter is a complex mixture of molecules, of varying carbon concentrations. A portion of this material is chromophoric and therefore exhibits the characteristic absorption coefficient exponentially decaying from ultraviolet to red. A portion of this chromophoric matter also fluoresces, generally in a broad peak centered on 460 nm in response to UV to blue wavelength excitation. Some distinct fluorophores have also been observed associated with specific proteins or amino acids (Coble 1996; Meyer et al. 1999). The CDOM fluorometer on the WET Labs triplet sensor is comprised of 370 nm LED and 460 nm detector pair, which detects the intensity

of the broad fluorescence peak found throughout fresh and marine water systems. This fluorescence signal is highly correlated the absorption coefficient at the excitation wavelength (Figure 4A and B) although there are two different slopes to the relationship, with the flatter slope associated with the wetland/bog stations (Figure 4B). This indicates that the concentration of fluorophores in that dissolved matter is lower than that found throughout the rest of the river system.

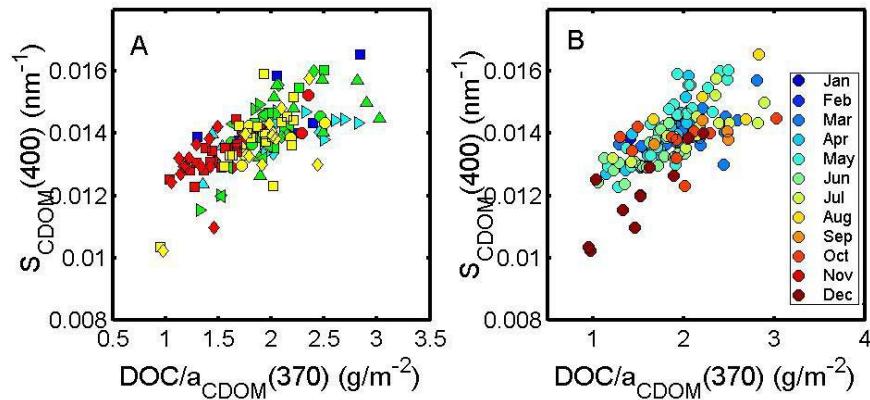


**Figure 4. Dissolved organic carbon (DOC) proxies. Relationship between in situ  $F_{CDOM}$  as measured with the WET Labs triplet sensor and the absorption of the dissolved matter at 370nm (the fluorescence excitation wavelength) measured spectrophotometrically on discrete samples collected during the monthly sampling program at all stations except the wetland drainage basins (A) and at the stations associated with wetland drainage basins (B). Optical proxies for DOC based upon the in situ  $F_{CDOM}$  (C) and dissolved absorption (D); symbols as in parts A and B.**

Dissolved organic carbon ranged from approximately 3 to 30 ( $\mu g\ C\ l^{-1}$ ). The absolute concentration of DOC was not seasonally dependent in the watershed but was certainly dependent upon station location. The highest concentrations consistently found in the tributaries draining wetland bogs (Sunkhaze Creek, and the Passadumkeag and Mattawamkeag Rivers). The lowest concentrations were found in the agricultural subwatershed drained by the Piscataquis River. The stations within the main branch of the Penobscot River varied depending upon location along the river and location of incoming tributaries, as expected. Within each station, seasonal dependence was observed with the highest concentrations occurring in the fall, lowest concentrations occurring in the spring during the two high discharge periods. In the spring, the discharge is a combination of snow and ice melt with precipitation

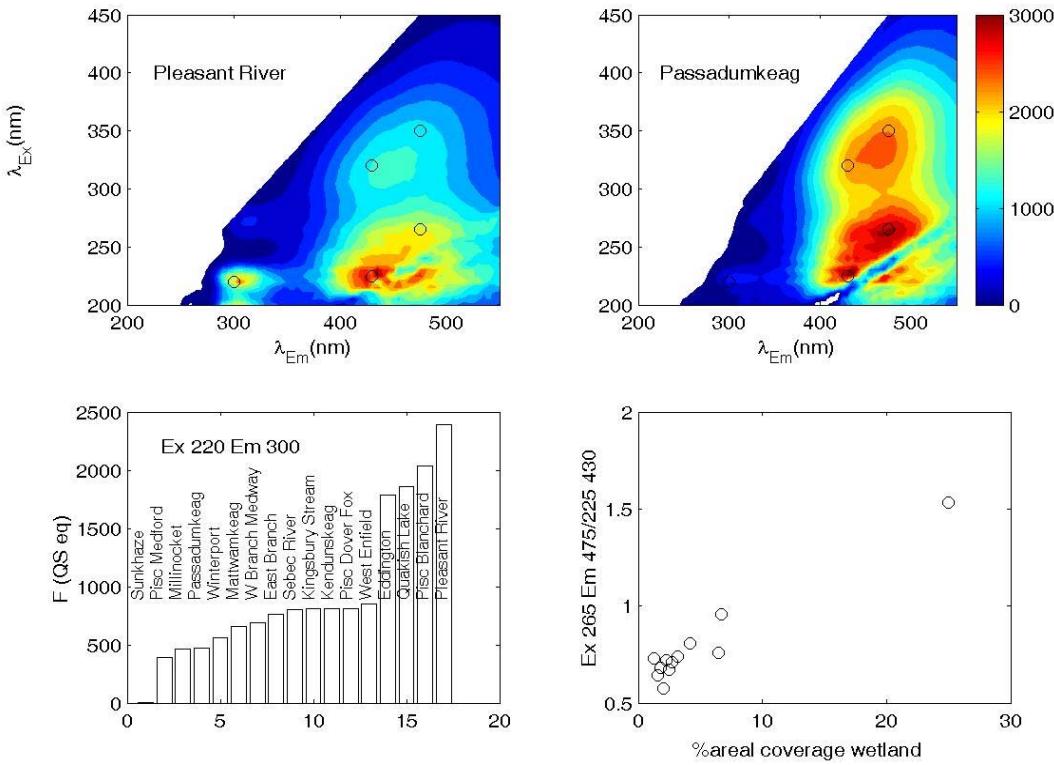
while the fall is due solely to precipitation. Thus the low DOC in the spring time is due to dilution of DOC-laden surface water by snow and ice. In addition, it is thought that the summer time low surface water period is a time of DOC production and accumulation. This material is released into the river system by fall precipitation, leading to the amplified DOC concentrations.

The relationship between  $F_{CDOM}$  and DOC is very similar to that observed with  $a_{CDOM}$  (Figure 4C) with two relationships. The flatter slope is associated with the high CDOM samples from wetland-draining tributaries, indicating that these waters have lower fluorescing efficiency or more non-fluorescing carbon-containing complexes, compared to the other sites. These relationships are robust throughout the year, exhibiting no seasonal dependence. Thus the in situ fluorescence proxy for DOC is robust but site sensitive, while the relationship between absorption and DOC is robust and insensitive to composition (either seasonally or by site). Interestingly the relationship between absorption and DOC is very robust and insensitive to station characteristics. Thus the ratio of colored matter to carbon-containing matter is constant in this environment (Figure 4D).



**Figure 5. Dissolved organic matter composition proxy based upon the exponential slope of the dissolved absorption spectrum,  $S$ , and the carbon containing fraction of the dissolved absorption coefficient ( $DOC/a_{CDOM}$ ); samples identified by station (A, symbol as in Figure 2A) and by sampling month (B).**

Dissolved Organic Matter Composition - One optical indicator of DOM composition is the exponential spectral slope of the absorption coefficient (Roesler et al. 1989; Carder et al. 1989). The exponential slope generally varies from 0.01 to 0.02, with lower slopes associated with newly formed labile organic matter, generally of marine origin and steeper slopes associated with old, refractory material, generally of freshwater origin (Carder et al. 1989; Simeon et al. 2003). The patterns observed here suggest that there are seasonal and location patterns in the spectral slope parameter (Figure 5A and B) although the slope is a good proxy for the carbon efficiency of the absorption. This means that the higher the slope of the absorption, the more carbon-rich is the material. In general the lower slope values were found associated with the high CDOM wetland bog sites while the highest values were found associated with the Piscataquis River and East Branch Penobscot in the far north of the state. The lowest values are observed at all sites in December (mean 0.012) and the highest values are observed at all sites in March (mean 0.015).



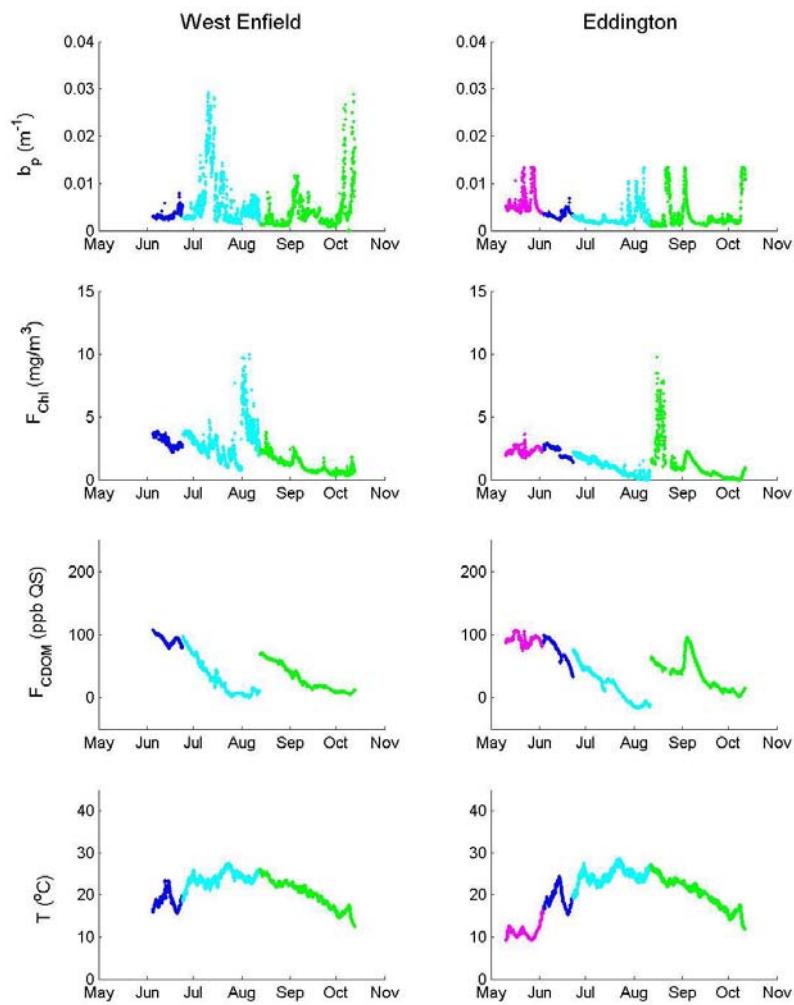
**Figure 6. Spectrofluorometric analysis of dissolved samples from July 2004. Examples of excitation/emission matrices for two sites typical of A) agricultural and B) wetland coverages. Symbols indicate spectral points where peaks were observed throughout the stations. C. Peak height (Ex 220, Em 300) for each station. Note the four highest values are located at stations downstream of watershed characterized by significant transformations. The next five stations are all associated with agricultural watersheds. Remaining stations are those found in the northern watersheds typical of forest and lumbered land cover. Thus this peak appears sensitive to CDOM breakdown products. D. Relation between peak ratio (Ex/Em 265/475 : 225/430) and the % areal coverage of the subwatershed by wetlands. A significant correlation is observed both with and without the high value point.**

Spectral fluorescence analysis provided more compositional information (Fig. 6). Two examples of 3d matrices are shown to demonstrate the distinct difference in peak occurrence and peak ratios as a function of watershed type (e.g. agricultural versus wetland). Fluorescence peaks have been found to be indicators of watershed type: stations located downstream of retention ponds where significant reworking of the CDOM is expected have strong peak at the ExEm 220/300 nm (Fig. 6C) while the stations downstream of wetlands have fluorescence peaks at ExEm 265/475 relative to 225/430 that are linearly related to the percent areal cover rage of wetland (Fig. 6D).

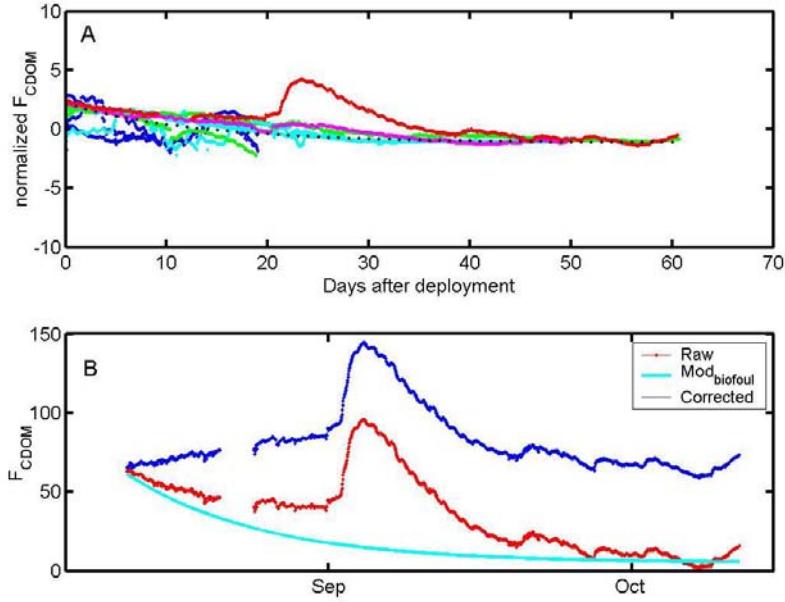
### Moored Optical Time Series

WET Labs Triplet sensors were deployed at four locations approximately May through October 2005 for one to three month intervals. The longest time series were obtained at two sites in the main branch of the Penobscot River at West Enfield and Eddington (Figure 7). The difference in observed values at

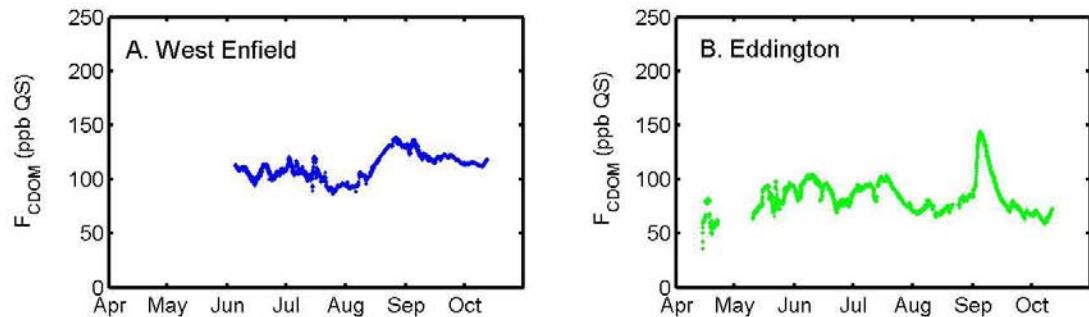
the end of one deployment relative to the beginning of the next deployment is an indication of sensor drift and/or biofouling. The only significant offsets were observed in the  $F_{CDOM}$  observations. A clear organic film that was observed on the recovered sensors was a significant absorber of both the outgoing excitation energy at 370 nm and the incoming emission energy at 460nm. It was observed, however, that if the observations from each deployment were normalized (differenced from mean and divided by variance) that the pattern in signal decay was remarkably constant between deployments and locations (Figure 8A). An exponential model was fit to that decay pattern, rescaled to the original observations (multiplied by variance and added to mean) and used to correct the original observations (Figure 8B). The result yielded time series of  $F_{CDOM}$  for which a background signal did not decay, there was no observed offset between deployments and in which there were distinct features associated with pulses of CDOM matter (Figure 9).



**Figure 7. Calibrated optical observations obtained from WET Labs triplet sensors deployed in the main branch Penobscot River at West Enfield (between the Piscataquis and Passadumkeag Rivers) and Eddington (between Sunkhaze Creek and Kenduskeag River). Each color represents a new sensor deployment. Only the  $F_{CDOM}$  displays significant offsets at the time of redeployment indicating significant sensor drift and/or biofouling. See text for details.**



**Figure 8.** A. Normalized  $F_{CDOM}$  time series for each deployment. Dotted black line indicates the average normalized signal to which an exponential model was fit. B. An example of  $F_{CDOM}$  biofouling correction procedure. Raw biofouled observation (red), biofouling signal (cyan), retrieved  $F_{CDOM}$  time series corrected for exponential decay in signal due to biofouling.



**Figure 9.** Corrected  $F_{CDOM}$  time series for West Enfield and Eddington.

## Carbon Time Series

In order to determine high-resolution carbon component time series we apply the conversion equations (proxy relationship) derived from the monthly sampling program to the in situ optical observations of  $F_{Chl}$ ,  $b_{bp}$ , and  $F_{CDOM}$ . These time series of concentrations are then converted to carbon loads by multiplying by the river discharge,  $D$  ( $ft^3/s$ ), time series provided by the US Geological Survey (Figure 9A, <http://waterdata.usgs.gov/me/nwis/current/>).

Thus we compute the algal carbon load from the calibrated  $F_{\text{Chl}}$  time series via:

$$C_{\text{algal}} (\text{kg C/h}) = F_{\text{Chl}} * C:\text{Chl} * D$$

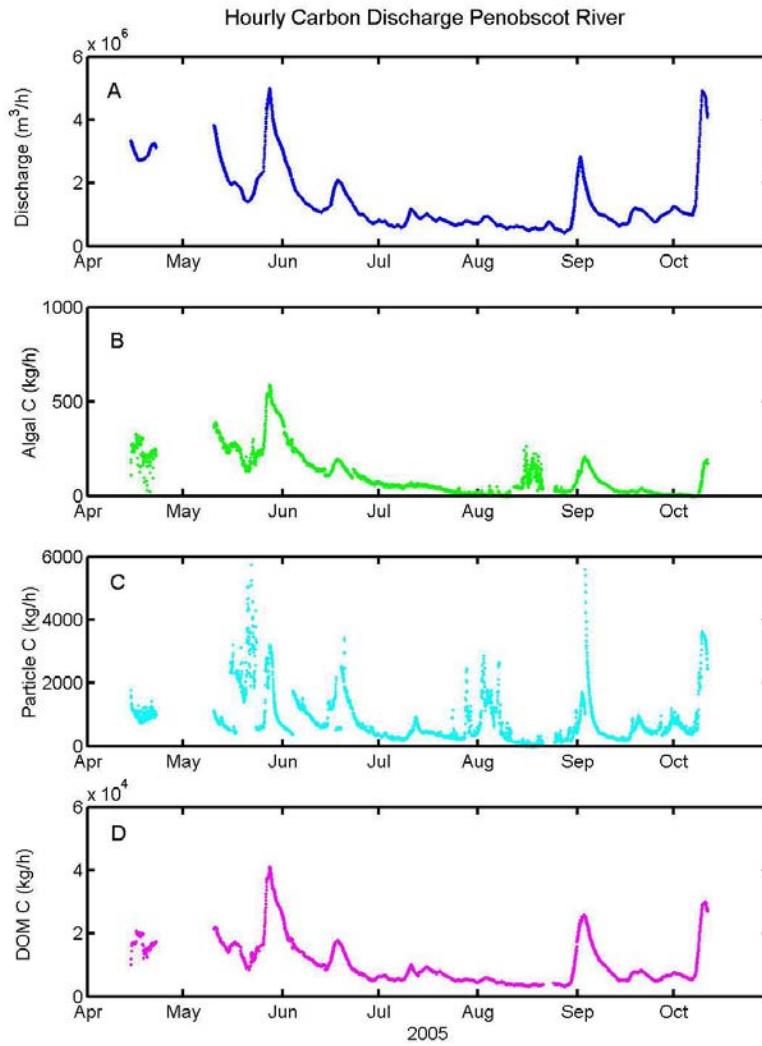
where  $F_{\text{chl}}$  is in units of ( $\text{mg Chl/m}^3$ ), an average carbon to chlorophyll ratio for phytoplankton of 50 ( $\text{g C/g Chl}$ ) (e.g. Cloern et al. 1995) and  $D$  is the discharge rate converted to ( $\text{m}^3/\text{h}$ ) with proper unit conversion. Similarly, the total particulate organic carbon load is computed from the time series  $b_{\text{bp}}$  observations using the proxy relationships for POC (Figure 3D) in which the low flow values are used until discharge exceeds  $2 \cdot 10^6$  ( $\text{m}^3/\text{h}$ ). Finally, the contribution of the algal carbon is removed so that the component is the non-algal particle carbon:

$$C_{\text{particles}} (\text{kg C/h}) = (b_{\text{bp}} - \text{offset})/\text{slope} * D - C_{\text{algal}}$$

where the conversions from  $\text{mg C/l}$  to  $\text{kg/m}^3$  are not explicit in the equation but were performed. Finally the carbon carried by the dissolved matter was computed from the  $F_{\text{CDOM}}$  time series observations using the proxy relationship for DOC (Figure 4C) with the appropriate unit conversions.

$$C_{\text{dissolved}} (\text{kg C/h}) = (F_{\text{CDOM}} - \text{offset})/\text{slope} * D .$$

These carbon time series (Figure 10) demonstrate that the amount of carbon being transported in the dissolved form from the terrestrial environment of the Penobscot Watershed to the marine system via Penobscot Bay dwarfs the contributions by algal or non-algal particles during the spring to fall interval, by two and one orders of magnitude, respectively. Integrating the time series over the season yields total carbon discharge into Penobscot Bay of 42.36 million  $\text{kg C}$  (42.36 thousand metric tons), partitioned as 38.94, 3.01, and 0.41 million  $\text{kg C}$  in dissolved, non-algal and algal components, respectively.



**Figure 10.** Time series of Penobscot River discharge (A) and organic carbon discharge for algal (B), non-algal particles (C) and dissolved matter (D) computed from carbon-specific optical proxy relationship applied to in situ moored optical observations. See text for details on computation.

## IMPACT/APPLICATIONS

The impact of terrestrial material on coastal waters takes a variety of roles. In this paper we have computed the input of algal and non-algal particles and dissolved matter in terms of organic carbon arising from the Penobscot River and emptying into the marine environment of Penobscot Bay and ultimately into the coastal current in the Gulf of Maine. The amount of carbon brought to the marine system is not negligible to the Gulf of Maine. The volume of freshwater that enters the GoM from the Penobscot River is approximately 2% of the volume of slope water that enters via the Northeast Channel (Brown and Irish 1993), but the concentration of organic carbon in the river water is more than 10 times the concentration found in the slope water, thus the Penobscot River contributes about 10% of the carbon that the slope water contributes. Approximately 90% of this organic carbon is in the dissolved fraction that is associated with strong absorption coefficients ( $>0.5 \text{ m}^{-1}$  in the blue region of

the spectrum), which contribute significantly to the surface ocean color. This colored matter has been shown to significantly impact retrieval of chlorophyll concentration from remotely sensed ocean color in general (Carder et al. 1989) and in the Gulf of Maine specifically (Yentsch and Phinney 1997; Sauer and Roesler, Ocean Optics Proceedings; Barnard et al. Ocean Optics Proceedings). While the total input of particulate organic carbon is not as significant as the dissolved fraction, the concentrations of this material are comparable or greater than those found in the marine waters and thus the river does act as a source of organic particles to the coastal surface waters. The fate of this material is the subject of ongoing investigations and modeling.

## RELATED PROJECTS

The NASA IDS program currently funds me with A. Barnard, B. Balch, G. Aiken, H. Xue, and T. Huntington to examine the ocean color remote sensing implications of river borne matter into Penobscot Bay.

The Gulf of Maine Ocean Observing System continues to provide operational optical time series for the coastal and basin waters of the Gulf of Maine. These observations provide the marine endmember optical properties against which the river observations can be placed in context.

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